Nonlinear polarization dynamics. I. The single-pulse equations

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We consider the polarization dynamics of a pulse propagating through an arbitrary nonlinear medium, in the limit of small nonlinearities, anisotropies, and dispersion, using the full SO(3) covariance of the Stokes parameters. The invariants of the motion are discovered and physically interpreted, a complete analogy with the "airplane-and-rotor" problem of rigid-body dynamics is established, and a full classification of the solutions for parity-invariant and non-parity-invariant media, with all propagation-axis rotation symmetries, is presented. In all cases, the problem is reduced to quadrature; in most cases, we find analytic solutions in terms of well-known functions.

I. INTRODUCTION

Maker, Terhune, and Savage[1] first demonstrated that the behavior of the polarization of a light beam propagating in a nonlinear medium can exhibit novel and interesting effects. They analyzed the self-interaction of a plane electromagnetic wave in a nonlinear, isotropic medium, and both predicted and observed that for elliptically polarized light the major axis of the ellipse rotates as a function of propagation length. They also pointed out that a nonlinear cubic crystal can exhibit an induced birefringence proportional to the intensity.

More recently, Kaplan[2,3] and Law[3] have studied the nonlinear interaction of two counterpropagating beams in an isotropic medium, and Yumoto and Otsuka[4] have considered such an interaction in a cubic crystal, where they find that a kind of "polarization chaos" results from the interaction. All of these authors have studied the polarization dynamics by the straightforward route of calculating how the electric field varies as a plane wave or waves propagate through the medium. In such an approach the desired result of the calculation—an understanding of how the polarization changes as the beam propagates—is often obscured since the state of the polarization is not always immediately obvious from the complex, slowly varying vector electric field amplitude that is calculated.

An alternate approach, which we develop in this paper, is to derive and work with the dynamical equations for the Stokes parameters of the light beam.[5] For a coherent beam there are three independent parameters, which are bilinear products of the vector components of the complex electric field amplitude. They are real, and contain all the information contained in that amplitude except the absolute phase of the field, which is usually not of interest; once the Stokes parameters are specified the polarization state of the beam can be immediately and easily visualized.

Stokes parameters have been used in studying the effects of stress fields on the propagation of light in a linear anisotropic medium,[6,7] and in examining the dynamics of polarization lasers.[8,9] And, indeed, Sala[10] and Gregori and Wabnitz[11] have used them already in studying the propagation of a plane wave through a nonlinear medium in the presence of dc field-induced birefringence. We considerably generalize and extend the Stokes-parameter formalism in this work by utilizing the SO(3) covariance of the Stokes parameters: They are components of a real, three-dimensional vector, and rotations in Stokes-vector space are homomorphic to SU(2) group transformations on the complex vector electric field amplitude. This allows us to relate the symmetries of the nonlinear medium directly to the symmetries of the equation of motion of the Stokes vector.

We do not consider the interesting effects of dc field-induced birefringence in this paper, but rather treat the more basic problem of a single beam, or pulse, propagating through a nonlinear medium. Previous work on this fundamental problem, which has been restricted to beam propagation, can be easily summarized: The propagation through an isotropic, parity-invariant medium has been considered,[1,10] as well as the propagation through a parity-invariant medium which has rotational symmetry C_4 about the axis of propagation.[11] In each case two invariants have been discovered, one of which corresponds to the intensity of the beam, and one of which has not been physically interpreted. Analytic solutions in terms of well-known functions have been given.

The use of the Stokes-vector formalism allows us to considerably extend this work. We consider parity-invariant *and* non-parity-invariant media, with the propagation axis characterized by no rotational symmetry or by rotational symmetry C_n, for *any* integer n (including $n=\infty$, or isotropy). We present a complete classification of the polarization equations of motion for all these cases, and prove that *in general* there are two invariants. We also discover the physical significance of the second invariant, which has not been realized even in the two instances where the existence of the invariant has been previously known. With the discovery of the general existence of two invariants, the solution of the polarization dynamics of a single beam, or pulse, in any anisotropic nonlinear medium can always be reduced to quadrature. But, by establishing more extensive analogies of polarization dynamics with rigid-body motion than have been developed to date,[10,11] we can do much more: If there is any rotational symmetry C_n about the propagation axis with $n>2$ we are able to construct an analytic solution in terms of well-known functions, whether the medium is parity invariant or not.

The plan of this paper is as follows. In Sec. II we briefly review the traditional approach of deriving equations of motion for the slowly varying electric field amplitudes. This defines our notation, and allows for comparison with the earlier work.[1-4] We do, however, generalize the usual treatment of a stationary plane wave to consider the propagation of a pulse. Although we neglect both loss and dispersion in the linear and self-induced anisotropies—assuming as usual that the time response of the susceptibility is so fast that the time-varying terms of Debye's equations[12] can be neglected—this is still an interesting extension. Since the intensity is a function of position in the pulse, and since the self-induced anisotropies are proportional to the intensity, the polarization evolves differently in different parts of the pulse, resulting from the full tensor form of self-phase-modulation. In Sec. III we introduce the Stokes parameters, derive the equation of motion for the Stokes vector, derive and physically interpret the invariants of the motion, and establish the correspondence between the coefficients in the equation of motion and the optical properties of the medium. In Sec. IV we establish analogies between the polarization dynamics in a nonlinear medium and rigid-body motion, which are useful both in visualizing the polarization dynamics and in constructing solutions of the equations. In Sec. V we consider the simplifications of the equation of motion that result if we make use of any crystal symmetries that may be present. Throughout, even in cases more complicated than those considered by earlier workers,[1-4,10,11] our equations take on a simpler form than in earlier formulations. We conclude with a short discussion in Sec. VI, deferring a detailed discussion of the new cases of physical interest presented here, and of the problem of two interacting beams, to upcoming communications.

II. THE FIELD AMPLITUDE EQUATIONS

To set the notation and make clear our assumptions, we begin by outlining the derivation of the electric field equations in the slowly-varying-amplitude approximation. The macroscopic polarization $\mathbf{P}(\mathbf{r},t)$ is given in terms of the macroscopic electric field $\mathbf{E}(\mathbf{r},t)$ by[12,13]

$$P_i(\mathbf{r},t)=\int dt'\,\chi_{ij}(t-t')E_j(\mathbf{r},t')+\int\int\int dt'\,dt''\,dt'''\,\chi^{(3)}_{ijkl}(t-t',t-t'',t-t''')E_j(\mathbf{r},t')E_k(\mathbf{r},t'')E_l(\mathbf{r},t''')\ , \tag{2.1}$$

where the subscripts denote Cartesian components and are to be summed over if repeated. The Fourier transforms of the susceptibilities,

$$\begin{aligned}\chi_{ij}(\omega)&=\int dt\,\chi_{ij}(t)e^{i\omega t}\ ,\\ \chi^{(3)}_{ijkl}(\omega',\omega'',\omega''')&=\int\int\int dt'\,dt''\,dt'''\,\chi^{(3)}_{ijkl}(t',t'',t''')\\ &\quad\times e^{i(\omega't'+\omega''t''+\omega'''t''')}\ ,\end{aligned} \tag{2.2}$$

which respectively characterize the linear and nonlinear response of the medium, satisfy

$$\begin{aligned}\chi_{ij}(\omega)&=\chi^*_{ij}(-\omega)\ ,\\ \chi^{(3)}_{ijkl}(\omega',\omega'',\omega''')&=\chi^{(3)*}_{ijkl}(-\omega',-\omega'',-\omega''')\ ,\end{aligned} \tag{2.3}$$

simply because the fields $\mathbf{E}(\mathbf{r},t)$ and $\mathbf{P}(\mathbf{r},t)$ are real. We further assume a lossless medium, and therefore also have[14,15]

$$\begin{aligned}\chi_{ij}(\omega)&=\chi^*_{ji}(\omega)\ ,\\ \chi^{(3)}_{ijkl}(\omega,\omega,-\omega)&=\chi^{(3)*}_{jilk}(\omega,\omega,-\omega)\ .\end{aligned} \tag{2.4}$$

It is then convenient to write the linear susceptibility $\chi_{ij}(\omega)$ as the sum of an isotropic and anisotropic part,

$$\chi_{ij}(\omega)=\chi(\omega)\delta_{ij}+\eta_{ij}(\omega)\ , \tag{2.5}$$

where $\chi(\omega)$ is one-third the trace of $\chi_{ij}(\omega)$ and is, by the first of Eqs. (2.4), real. The traceless matrix $\eta_{ij}(\omega)$ (which is Hermitian by the same equation), describes the linear anisotropy of the medium. That anisotropy can include both birefringence and optical activity, the latter of which has not been considered in treatments of the single beam problem.[10,11] We assume that the linear anisotropy, as well as the nonlinearities, are small in magnitude. That is,

$$|\eta_{ij}|,\ |\chi^{(3)}_{ijkl}E^2|\ll 1\ , \tag{2.6}$$

where E is a typical electric field amplitude.

In considering the propagation of light through the medium, earlier workers[1-4,10,11] have assumed that the electric field is a plane wave (or plane waves) with an amplitude which may be slowly varying in space, but is constant in time,

$$\mathbf{E}(\mathbf{r},t)=\mathbf{e}(z)e^{i(k_0z-\omega_0t)}+\text{c.c.}\ , \tag{2.7}$$

where z is chosen as the propagation direction. We give a more general discussion here, to consider pulse propagation; we write, instead of Eq. (2.7),

$$\mathbf{E}(\mathbf{r},t)=\mathbf{e}(z,t)e^{i(k_0z-\omega_0t)}+\text{c.c.}\ , \tag{2.8}$$

where $\mathbf{e}(z,t)$ is now supposed to vary significantly over distances much greater than k_0^{-1}, and over times much greater than ω_0^{-1}, and we ignore any transverse spatial dependence of the beam. Putting Eqs. (2.1) and (2.8) in Maxwell's equations, we neglect any dispersion in the nonlinearities and anisotropies, setting

$$\eta_{ij}(\omega)=\eta_{ij}(\omega_0)\equiv\eta_{ij} ,$$
$$\chi^{(3)}_{ijkl}(\omega,\omega,-\omega)=\chi^{(3)}_{ijkl}(\omega_0,\omega_0,-\omega_0)\equiv\chi^{(3)}_{ijkl} . \quad (2.9)$$

However, we include to first-order dispersion in the (in general larger) isotropic part of the susceptibility, setting [cf. Eq. (2.5)]

$$\chi_{ij}(\omega)=[\chi(\omega_0)+(\omega-\omega_0)\chi'(\omega_0)]\delta_{ij}+\eta_{ij}(\omega_0) , \quad (2.10)$$

where $\chi'(\omega)=d\chi(\omega)/d\omega$. We find

$$\frac{\partial e_i}{\partial z}+\frac{1}{v}\frac{\partial e_i}{\partial t}=\frac{2\pi i k_0}{\epsilon_0}(\eta_{ij}e_j+3\chi^{(3)}_{ijkl}e_j e_k e_l^*) , \quad (2.11)$$

where the indices run over (x,y); it is easy to verify that, under the assumptions (2.6), e_z is negligible and we henceforth neglect it. As usual the dielectric constant ϵ_0 is

$$\epsilon_0=1+4\pi\chi(\omega_0) , \quad (2.12)$$

the carrier wave number and frequency are related by $k_0^2c^2=\omega_0^2\epsilon_0$, and

$$v=\frac{d\omega_0}{dk_0} \quad (2.13)$$

is the group velocity at the carrier frequency. In deriving Eq. (2.11) we have neglected any dc or harmonic field generation by the pulse, and have used the fact that $\chi^{(3)}_{ijkl}$, Eq. (2.9), is symmetric under exchange of j and k ("intrinsic permutation symmetry"[14,15]) and, by virtue of Eq. (2.4), under exchange of i and l. Finally, we make a change of independent variables to the following dimensionless coordinates:

$$\xi=k_0(z-vt) ,$$
$$\tau=k_0vt . \quad (2.14)$$

Here ξ is a dimensionless spatial coordinate in a reference frame following the pulse at its group velocity v; τ is the dimensionless time variable in this pulse reference frame. Equations (2.11) then become

$$\dot{e}_i=\frac{2\pi i}{\epsilon_0}(\eta_{ij}e_j+3\chi^{(3)}_{ijkl}e_je_ke_l^*) \;\; (i,j,k,l=x,y) , \quad (2.15)$$

where the overdot denotes a partial derivative with respect to the dimensionless time variable τ. Since at an initial time $\tau=0$ we have $e_j=e_j(\xi)$ varying through the pulse, Eq. (2.15) shows that, within our approximations, e_j will evolve "locally" at each point within the pulse; the $\chi^{(3)}$ contribution thus describes the tensor form of self-phase-modulation. Note that in making the approximation (2.10) we neglect group-velocity dispersion, which in a pulse-compression experiment would also be an important effect. It is to the determination of the electromagnetic field specified by Eqs. (2.15) that the rest of this paper is devoted. We may recover the equations of earlier workers,[1–4,10,11] who considered the simpler problem of a stationary plane wave (2.7): Repeating the analysis for that case we find Eq. (2.11) without the time derivative, and we again find Eqs. (2.15), but with the overdot now indicating differentiation with respect to k_0z and the e_j, being constant in time, varying along the direction in which the stationary beam is propagating. Of course, those earlier workers considered only the fairly simple form Eqs. (2.15) take for parity invariant media which are isotropic,[1–3,10] or for which propagation occurs in a very high symmetry direction.[4,11] We consider the most general form (2.15) here.

III. THE STOKES-VECTOR EQUATIONS

For a transverse wave the amplitude $\mathbf{e}$ appearing in Eqs. (2.15) can be written as a complex vector of dimension two,

$$\begin{bmatrix} e_x \\ e_y \end{bmatrix} \equiv \begin{bmatrix} e_1 \\ e_2 \end{bmatrix} . \quad (3.1)$$

In this space the basis vectors, which correspond to polarization eigenstates, transform into one another by SU(2) rotations. Thus in a formal sense the polarization dynamics can be thought of as an effective spin-$\frac{1}{2}$ system. However, working directly with Eqs. (2.15) for e_i has certain disadvantages: The e_i themselves are complex, as are the η_{ij} and $\chi^{(3)}_{ijkl}$. And those latter quantities satisfy certain constraints [Eqs. (2.3) and Eqs. (2.4) in a lossless medium] even before any material symmetries are taken into account. We[8,9] and others[6,7,10,11] find it more useful to work with bilinear combinations of the complex electric field. These quantities are real, the equations they satisfy involve only real quantities for a material of arbitrary symmetry, and they completely specify the electric field except for its absolute phase, which is usually not of interest. The bilinear combinations are elements of the direct product of the above mentioned effective spin-$\frac{1}{2}$ system with itself, and such a direct product transforms irreducibly as a spin-1 and a spin-0 representation,

$$\tfrac{1}{2}\otimes\tfrac{1}{2}=1\oplus 0 . \quad (3.2)$$

SU(2) transformations of the spin-1 representation are homomorphic to SO(3) rotations of a vector: the Stokes vector. The spin-0 representation is invariant under SU(2) transformations and thus under the SO(3) rotations; we see below that it corresponds simply to the magnitude of the Stokes vector.

We define the Stokes parameters by one[8] of the standard conventions,

$$s_0=e_j^*(\sigma_0)_{jk}e_k=e_j^*e_j ,$$
$$s_i=e_j^*(\sigma_i)_{jk}e_k, \;\; i=1,2,3 . \quad (3.3)$$

Here σ_0 is the unit 2×2 matrix, and the σ_i are the Pauli spin matrices. The sum over j and k in Eq. (3.3), and corresponding sums over repeated indices which appear in this section, range only over 1 and 2. The parameters s_0 and s_i are all easily seen to be real, and a little algebra reveals the identity

$$s_0^2=\mathbf{s}\cdot\mathbf{s} , \quad (3.4)$$

where we have introduced three unit vectors in "Stokes-vector space" to allow us to introduce a Stokes vector $\mathbf{s}$; the transformation of the Pauli spin matrices under SU(2) guarantees that such transformations indeed correspond to rotations of **s**. From Eq. (3.4) we see that there are three independent Stokes parameters for a coherent beam: they correspond to the three physical variables necessary to define the polarization and intensity of a coherent beam.[5] From the first of Eqs. (3.3) we see that s_0 is proportional to the intensity of the beam.

The correspondence between the Stokes vector **s** and the polarization is easiest seen using the Poincaré sphere, a surface in Stokes-vector space (see Fig. 1). It is centered at the origin and each point on it, which specifies a direction $\hat{\mathbf{s}}$, identifies a polarization state. From Eq. (3.3) it is easy to verify that all vectors **s** lying in the (1,3) plane represent linearly polarized light; rotating through an angle θ in the (1,3) plane corresponds to rotating the plane of polarization by $\theta/2$ in real space. The directions $\hat{2}$ and $-\hat{2}$ correspond to left- and right-handed circular polarizations, respectively. Remaining directions on the upper and lower hemispheres correspond to left- and right-handed elliptical polarizations; moreover, opposing directions of **s** ($\pm\mathbf{s}$) correspond to orthogonal polarizations.

We can now deduce an equation of motion for the Stokes vector. From the definition (3.3) we obtain

$$\begin{aligned}\dot{s}_i &= \dot{e}_j^*(\sigma_i)_{jk}e_k + e_j^*(\sigma_i)_{jk}\dot{e}_k \\ &= 2\,\mathrm{Re}[e_j^*(\sigma_i)_{jk}\dot{e}_k]\ ,\end{aligned} \tag{3.5}$$

where in the second of Eqs. (3.5) we have used the Hermiticity of the Pauli spin matrices. Substituting Eq. (2.15) into Eq. (3.5) and making use of the Pauli spin matrix identities,

$$\begin{aligned}&(\sigma_i)^*_{lm}(\sigma_i)_{np} + \delta_{lm}\delta_{np} = 2\delta_{\ln}\delta_{mp}\ ,\\ &\sigma_i\sigma_j = \sigma_0\delta_{ij} + i\epsilon_{ijk}\sigma_k\end{aligned} \tag{3.6}$$

as well as the symmetry relations (2.4) we find

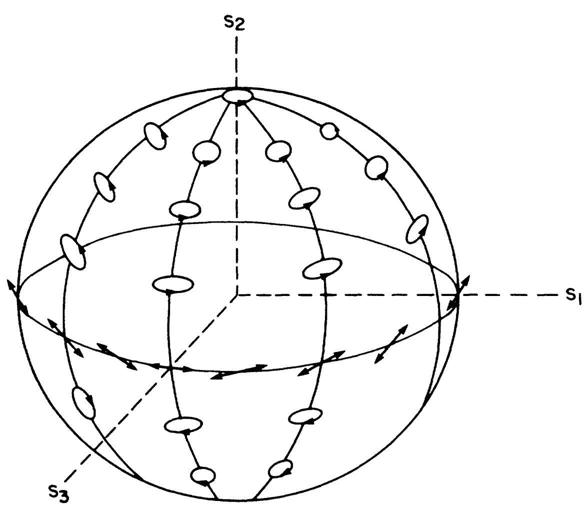

FIG. 1. The Poincaré sphere, showing the correspondence between polarization and the directions of the Stokes vector.

$$\dot{s}_i = \epsilon_{ijk}s_j(v_k + s_0u_k + w_{kl}s_l)\ , \tag{3.7}$$

where the material coefficients are defined as

$$\begin{aligned}v_k &= \left[\frac{2\pi}{\epsilon_0}\right](\sigma_k)_{ji}\eta_{ij}\ ,\\ u_k &= \left[\frac{3\pi}{\epsilon_0}\right](\sigma_k)_{ji}\chi^{(3)}_{ijll}\ ,\\ w_{kl} &= \left[\frac{3\pi}{\epsilon_0}\right](\sigma_k)_{ji}\chi^{(3)}_{ijmn}(\sigma_l)_{mn}\ ,\end{aligned} \tag{3.8}$$

and ϵ_{ijk} is the Levi-Civita symbol, the totally antisymmetric third-rank SO(3) tensor. Using the Hermiticity of the Pauli spin matrices, the symmetry relations (2.4), and the intrinsic permutation symmetry of the third-order susceptibility one can show that the coefficients (3.8) are all real and that w_{kl} is symmetric,

$$w_{kl} = w_{lk}\ . \tag{3.9}$$

The transformation of the Pauli spin matrices under SU(2) guarantee that, under the homomorphic SO(3) rotations in Stokes-vector space, the v_k and u_k transform like the Cartesian components of vectors, and w_{kl} like the Cartesian components of a second-rank tensor. Thus we are justified in writing Eqs. (3.7) in vector notation,

$$\dot{\mathbf{s}} = \mathbf{s}\times(\mathbf{v} + s_0\mathbf{u} + \overleftrightarrow{\mathbf{w}}\cdot\mathbf{s})\ . \tag{3.10}$$

Recall that, for a pulse, the dot indicates the (dimensionless) time derivative at a given position in the reference frame moving with the pulse; for a stationary wave it indicates the (dimensionless) spatial derivative in the direction of propagation of the beam. The real Eqs. (3.10) completely describe the polarization dynamics of a single pulse or beam in a nonlinear medium of arbitrary symmetry, under the assumptions given in Sec. II, and have not been written down before. We consider their solution, for arbitrary material symmetry about the propagation axis, in Sec. V; we first identify the physical significance of the material tensors (3.8).

The first term on the right-hand side of Eq. (3.10) represents linear anisotropy, since it leads to a precession of the Stokes vector about the direction $\hat{\mathbf{v}}$ (see Fig. 1) at a rate independent of the intensity [see also the first of Eqs. (3.8)]. The second term, which is proportional to the intensity, represents induced anisotropies and the third-term higher-order, nonlinear symmetry effects. Now, our particular choice of definition of the Stokes vector (3.3) corresponds to writing the electric field in an (x,y) linear polarization basis. Such a choice is arbitrary and does not have any physical significance, but once it is made we can identify the physical effects associated with the components of **v**, **u**, and $\overleftrightarrow{\mathbf{w}}$ by recalling that the directions $\hat{1}$ and $\hat{3}$ in Stokes-vector space correspond to linear polarization, and the direction $\hat{2}$ to circular polarization:

$$\begin{aligned}
&v_1 \to \text{birefringence} , \\
&v_2 \to \text{optical activity} , \\
&v_3 \to \text{birefringence} ; \\
&u_1 \to \text{self-induced birefringence} , \\
&u_2 \to \text{self-induced optical activity} , \\
&u_3 \to \text{self-induced birefringence} ; \\
&w_{11}, w_{13}, w_{33} \to \text{linear-linear polarization interaction} , \\
&w_{12}, w_{23} \to \text{linear-circular polarization interaction} , \\
&w_{22} \to \text{circular-circular polarization interaction} .
\end{aligned} \tag{3.11}$$

Note that w_{kl}, being a second-rank symmetric tensor, consists of a spin-2 and a spin-0 component. The spin-0 component, proportional to the trace w_{kk}, does not contribute to the polarization dynamics (3.10); thus there are really only five independent components of physical significance.

Next, we turn to the invariants of the motion. Our discussion will deal with a pulse of radiation, but can be easily simplified to treat the stationary wave case. From Eq. (3.10) it is easy to verify that two invariants of the motion are

$$s_0, \ \tfrac{1}{2} s_k w_{kl} s_l + s_0 \mathbf{u}\cdot\mathbf{s} + \mathbf{v}\cdot\mathbf{s} . \tag{3.12}$$

These invariants are constant in time τ, but are in general a function of ξ as determined by the initial pulse at $\tau=0$. In particular for s_0, which is simply proportional to the intensity, we have

$$s_0(\xi,\tau) = s_0(\xi,0) , \tag{3.13}$$

and the intensity propagates at the group velocity with the initial shape of the pulse.

The existence of a second invariant has been noted for propagation along certain high symmetry directions in a parity-invariant medium.[10,11] But in establishing the second of Eqs. (3.12) as an invariant from Eqs. (3.10), we present a new result: Such a second invariant generally exists in a lossless medium, subject only to the approximations leading to Eqs. (2.15), regardless of the symmetry. This second invariant (3.12) is determined not only by the initial intensity but by the initial polarization, which of course can also vary through the pulse at $\tau=0$. And even in the simple cases where its existence has been noted,[10,11] its physical significance has not been established. This we now proceed to do, for the most general lossless medium. We recall from thermodynamics that the work per unit volume done to polarize a medium by subjecting it to an increasing electric field from external sources is given by[16]

$$dW = -\mathbf{P}\cdot d\mathbf{E} , \tag{3.14}$$

where $\mathbf{P}$ must be specified as a function of $\mathbf{E}$ to calculate the total work. To do this, we imagine we have

$$\begin{aligned}
E_j(\mathbf{r},t) &= \overline{E}_j(\mathbf{r},t) + \text{c.c.} , \\
P_j(r,t) &= \overline{P}_j(\mathbf{r},t) + \text{c.c.} ,
\end{aligned} \tag{3.15}$$

where the $\overline{E}_j$ and $\overline{P}_j$ contain only positive-frequency parts. Here $E_j(\mathbf{r},t)$ and $P_j(\mathbf{r},t)$ are equal to our actual fields at times close to t, but are adiabatically brought up to those values from zero at $t=-\infty$. For slowly varying amplitudes, the time-averaged work is then[13]

$$dW = -2\,\text{Re}[\overline{P}_j^*(\mathbf{r},t) d\overline{E}_j(\mathbf{r},t)] \tag{3.16}$$

and, since at constant temperature the integral of this can be identified as the (time average of) a free-energy density,[13,16] we have

$$\begin{aligned}
\langle F \rangle &= -2\,\text{Re} \int_0^{\mathbf{E}(\mathbf{r},t)} \overline{P}_j^*(\mathbf{r},t) d\overline{E}_j(\mathbf{r},t) \\
&= -2\,\text{Re} \int_{-\infty}^{t} \overline{P}_j^*(\mathbf{r},t) \frac{\partial \overline{E}_j(\mathbf{r},t)}{\partial t} dt .
\end{aligned} \tag{3.17}$$

We can now specify $\overline{P}_i(\mathbf{r},t)$ in terms of $\overline{E}_j(\mathbf{r},t)$ by using Eqs. (2.8) and (2.10) in Eq. (2.1). We find, neglecting any harmonic generation,

$$\overline{P}_i(\mathbf{r},t) = \chi_{ij}(\omega_0)\overline{E}_j(\mathbf{r},t) - \omega_0 \chi'(\omega_0)\overline{E}_i(\mathbf{r},t) + i \frac{\partial \overline{E}_i(\mathbf{r},t)}{\partial t}\chi'(\omega_0) + 3\chi^{(3)}_{ijkl}(\omega_0,\omega_0,-\omega_0)\overline{E}_j(\mathbf{r},t)\overline{E}_k(\mathbf{r},t)\overline{E}_l^*(\mathbf{r},t) . \tag{3.18}$$

Putting Eqs. (3.18) into Eq. (3.17), performing the integrals and using Eq. (2.8), we insert Kronecker deltas in the resulting contractions and use the Pauli spin matrix identities (3.6) to obtain

$$\begin{aligned}
\langle F \rangle = &-[\chi(\omega_0) - \omega_0\chi'(\omega_0)]s_0 - \left[\frac{\epsilon_0}{8\pi}\right] w_{kk} s_0^2 \\
&- \left[\frac{\epsilon_0}{8\pi}\right] (\tfrac{1}{2} s_k w_{kl} s_l + s_0 \mathbf{u}\cdot\mathbf{s} + \mathbf{v}\cdot\mathbf{s}) .
\end{aligned} \tag{3.19}$$

This free-energy density is a function only of the two invariants (3.12), and hence at each ξ is constant in the time τ. Alternately, we can identify the second invariant (3.12) as a simple function of the intensity and the free-energy density. For convenience we refer to it as the "free-energy invariant."

We close this section by noting that an alternate scheme for calculating the polarization work in thermodynamics is to exclude the energy of interaction with the external field and calculate the net work necessary to polarize in zero field.[16] Instead of Eq. (3.14) one finds $dW = \mathbf{E}\cdot d\mathbf{P}$, and the resulting expression for fields of the form (3.18) is

$$2\,\mathrm{Re}\int_0^{\mathbf{P}(\mathbf{r},t)} \bar{E}_j^*(\mathbf{r},t)d\bar{P}_j(\mathbf{r},t)$$
$$= [\chi(\omega_0)+\omega_0\chi'(\omega_0)]s_0$$
$$+\frac{\epsilon_0}{4\pi}(\tfrac{3}{2}s_k w_{kl}s_l+\tfrac{3}{2}w_{ll}s_0^2+3s_0\mathbf{u}\cdot\mathbf{s}+\mathbf{v}\cdot\mathbf{s}) \ . \quad (3.20)$$

Perhaps not surprisingly, this expression is not just a function of the invariants, and hence is not constant in the time τ. We see in Sec. V that Eq. (3.10) typically has periodic solutions, so the expression in Eq. (3.20) oscillates periodically in τ.

IV. RIGID-BODY ANALOGIES

It has been pointed out by Sala[10] and Gregori and Wabnitz[11] that the equations of motion for the Stokes parameters are similar in form to the equations that appear in the theory of rigid-body motion; this is not surprising, since the Stokes parameters are related to the angular momentum of the electromagnetic field.[17] The equation of motion (3.10) we have derived here for the Stokes vector is more general than those considered by other workers,[10,11] but it is still possible to find analogies with rigid-body motion. To do this, we recall three problems in classical mechanics. The first two problems are well known, but we present them here for reference, and to set the notation for the third problem. These analogies are interesting because they demonstrate previously unappreciated formal similarities between quite different physical problems. But they also have important practical consequences: On the basis of earlier work in rigid-body motion, we are able in Sec. V to establish analytic solutions in terms of well-known functions for almost all of the problems involving the polarization dynamics of a single pulse or beam.

We first consider a symmetric rigid body[18] with a gyromagnetic ratio Γ relating the magnetic dipole moment $\mathbf{M}$ of the body to its angular momentum $\mathbf{L}$,

$$\mathbf{M}=\Gamma\mathbf{L} \ . \quad (4.1)$$

If this body is placed in a magnetic field $\mathbf{B}$ the torque on it is $\mathbf{M}\times\mathbf{B}$, so the equation of motion is

$$\frac{d\mathbf{L}}{dt}=\Gamma\mathbf{L}\times\mathbf{B} \ , \quad (4.2)$$

the Bloch equation familiar from nuclear magnetic resonance[19] and quantum optics.[20]

Next, we consider a rigid-body rotating in free space. Writing the angular momentum in an inertial frame as

$$\mathbf{L}=L_i\hat{\mathbf{e}}_i=L_i'\hat{\mathbf{e}}_i' \ , \quad (4.3)$$

where the sum in Eq. (4.3) ranges from 1 to 3, and the unit vectors $\hat{\mathbf{e}}_i$ specify the inertial frame, while the $\hat{\mathbf{e}}_i'$ are "attached" to the body. Defining as usual the "space" and "body" derivatives of the angular momentum,

$$\left[\frac{d\mathbf{L}}{dt}\right]_{\text{space}}=\hat{\mathbf{e}}_i\frac{dL_i}{dt} \ ,$$
$$\left[\frac{d\mathbf{L}}{dt}\right]_{\text{body}}=\hat{\mathbf{e}}_i'\frac{dL_i'}{dt} \ , \quad (4.4)$$

[where, of course, the derivative in Eq. (4.2) is a space derivative], we find[21]

$$\left[\frac{d\mathbf{L}}{dt}\right]_{\text{space}}=\left[\frac{d\mathbf{L}}{dt}\right]_{\text{body}}+\boldsymbol{\omega}\times\mathbf{L} \ , \quad (4.5)$$

where $\boldsymbol{\omega}$ is the angular velocity of the body in the inertial frame. The angular momentum is related to the angular velocity by the moment of inertia tensor whose components I_{ij} in the body frame,

$$L_i'=I_{ij}\omega_j' \ , \quad (4.6)$$

are time independent. Thus the equation of motion for the body,

$$\left[\frac{d\mathbf{L}}{dt}\right]_{\text{space}}=0 \ , \quad (4.7)$$

leads, using Eqs. (4.5) and (4.6), to

$$\left[\frac{d\mathbf{L}}{dt}\right]_{\text{body}}=-(\overleftrightarrow{\mathbf{I}}^{-1}\cdot\mathbf{L})\times\mathbf{L} \ . \quad (4.8)$$

If the body axes are chosen so the moment of inertia tensor is diagonal, we recover Euler's equations of motion for a rigid body,

$$\dot{L}_1'=-L_2'L_3'(I_2^{-1}-I_3^{-1}) \ ,$$
$$\dot{L}_2'=-L_3'L_1'(I_3^{-1}-I_1^{-1}) \ , \quad (4.9)$$
$$\dot{L}_3'=-L_1'L_2'(I_1^{-1}-I_2^{-1}) \ ,$$

where I_i are the principal moments of inertia, and we have written the equations in terms of $\dot{L}_i'$ instead of the more usual form for $\dot{\omega}_i'$.

Finally, we consider the mechanical system shown in Fig. 2. Rigid body 1 is hollow and in general unsymmetric, having unequal principal moments of inertia. It includes, as in the figure, a rigidly fixed rod upon which is mounted rigid body 2, a homogeneous sphere (or at least with one principal axis along the rod). There are supposed to be no net torques on the combined system. We imagine the second body is very rapidly rotating about the rod, so that is is not a bad approximation to assume that essentially all of its angular momentum is in the direction

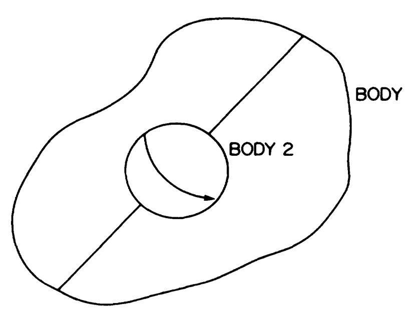

FIG. 2. Rigid-body analogy for the Stokes-vector equation of motion: the "airplane-and-rotor" problem.

of the rod, and constant when referred onto a reference frame attached to body 1. That is,

$$\left[\frac{d\mathbf{L}_2}{dt}\right]_{\text{body}}=0\ , \tag{4.10}$$

where the subscript body refers to body 1. This is a common approximation made, for example, in considering the dynamics of a rotor in an airplane.[22] Now

$$\left[\frac{d\mathbf{L}_2}{dt}\right]_{\text{space}}=\left[\frac{d\mathbf{L}_2}{dt}\right]_{\text{body}}+\boldsymbol{\omega}_1\times\mathbf{L}_2=\mathbf{N}_2\ , \tag{4.11}$$

where ω_1 is the angular velocity of body 1, and $\mathbf{N}_2$ is the torque exerted on body 2 by body 1. The corresponding equation for body 1 is

$$\left[\frac{d\mathbf{L}_1}{dt}\right]_{\text{space}}=\left[\frac{d\mathbf{L}_1}{dt}\right]_{\text{body}}+\boldsymbol{\omega}_1\times\mathbf{L}_1=\mathbf{N}_1\ , \tag{4.12}$$

where $\mathbf{N}_1=-\mathbf{N}_2$ since there is no net torque on the system. Using this with Eqs. (4.10)–(4.12) we find

$$\left[\frac{d\mathbf{L}_1}{dt}\right]_{\text{body}}=-\boldsymbol{\omega}_1\times\mathbf{L}\ , \tag{4.13}$$

where $\mathbf{L}=\mathbf{L}_1+\mathbf{L}_2$ is the total angular momentum of the system. Writing

$$\boldsymbol{\omega}_1=\overleftrightarrow{\mathbf{I}}_1^{-1}\cdot\mathbf{L}_1=\overleftrightarrow{\mathbf{I}}_1^{-1}\cdot\mathbf{L}-\overleftrightarrow{\mathbf{I}}_1^{-1}\cdot\mathbf{L}_2\ , \tag{4.14}$$

and using Eq. (4.10) again we can write (4.13) as

$$\left[\frac{d\mathbf{L}}{dt}\right]_{\text{body}}=\mathbf{b}\times\mathbf{L}-\boldsymbol{\Omega}(\mathbf{L})\times\mathbf{L}\ , \tag{4.15}$$

where

$$\mathbf{b}=\overleftrightarrow{\mathbf{I}}_1^{-1}\cdot\mathbf{L}_2,\quad \boldsymbol{\Omega}(\mathbf{L})=\overleftrightarrow{\mathbf{I}}_1^{-1}\cdot\mathbf{L}\ . \tag{4.16}$$

This is a closed equation for the total angular momentum of the system, referred to axes attached to body 1, under the approximation (4.10); it is really just a restatement of the "airplane-and-rotor" result (4.13). However, note that the vector $\mathbf{b}$ is constant since we have assumed (4.10) and, with respect to axes attached to body 1, $\overleftrightarrow{\mathbf{I}}_1$ is constant.

Returning now to the equations (3.10) for the Stokes vector, we introduce

$$\mathbf{b}\equiv-(\mathbf{v}+\mathbf{u}s_0),\quad \boldsymbol{\Omega}(\mathbf{s})\equiv\overleftrightarrow{\mathbf{I}}^{-1}\cdot\mathbf{s}\ , \tag{4.17}$$

where we define

$$I_{ij}^{-1}\equiv w_{ij}\ , \tag{4.18}$$

The $\mathbf{b}$ of equation (4.17) is a constant vector, since $\mathbf{v}$ and $\mathbf{u}$ are constant (as are $\overleftrightarrow{\mathbf{w}}$ and thus $\overleftrightarrow{\mathbf{I}}$), and s_0 is a constant of the motion. Using Eqs. (4.17), Eq. (3.10) may be written as

$$\dot{\mathbf{s}}=\mathbf{b}\times\mathbf{s}-\boldsymbol{\Omega}(\mathbf{s})\times\mathbf{s}\ , \tag{4.19}$$

and, referring back to Eq. (4.15), we see that the dynamics of the Stokes vector in a nonlinear medium is, in general, equivalent to the dynamics of the total angular momentum of an airplane and rotor, referred to axes attached to the airplane, under the usual approximation (4.10) made in that problem. Of course, since $\overleftrightarrow{\mathbf{w}}$ is a symmetric tensor we can always find a reference frame in Stokes-vector space in which it is diagonal,

$$\overleftrightarrow{\mathbf{w}}=\begin{bmatrix} I_1^{-1} & 0 & 0 \\ 0 & I_2^{-1} & 0 \\ 0 & 0 & I_3^{-1}\end{bmatrix}\ . \tag{4.20}$$

However, there is no guarantee that the diagonal elements will be positive. Nonetheless, because the polarization dynamics (3.10) are insensitive to the trace of $\overleftrightarrow{\mathbf{w}}$, we can always add a large enough constant to all the I_i^{-1} to make the diagonal elements of Eq. (4.20) positive, and preserve the analogy with rigid-body motion. Note that while it is the moment of inertia tensor elements that are analogous to the coefficients of polarization interaction Eq. (4.18), it is the angular momentum of the rotor (through the moment of inertia tensor) that plays the role of the (linear and self-induced) anisotropy [Eqs. (4.16) and (4.17)].

Of course, if some of the material tensors $\mathbf{u},\mathbf{v},\overleftrightarrow{\mathbf{w}}$ vanish or take a simple form, simpler analogies result, as have been noted by other authors.[7,10,11] The absence of linear and self-induced anisotropies [$\mathbf{b}=0$ in Eq. (4.17)] is equivalent to removing the rotor [$\mathbf{L}_2=0$ in Eq. (4.16)]. Then the Stokes-vector equation (4.19) reduces to

$$\begin{aligned}\dot{\mathbf{s}}&=-\boldsymbol{\Omega}(\mathbf{s})\times\mathbf{s}\\ &=-(\overleftrightarrow{\mathbf{I}}^{-1}\cdot\mathbf{s})\times\mathbf{s}\ ,\end{aligned} \tag{4.21}$$

and the analogy is with the torque-free motion of a rigid body [the airplane—see Eq. (4.8), and Eq. (4.15) with $\mathbf{b}=0$]. Finally, in the absence of nonlinearities—and even in some special cases where they are present [see Eq. (5.11)]—the dynamics (4.19) reduces to a simple Bloch equation (4.2) with a constant magnetic field.

V. SYMMETRIES AND SOLUTIONS

We now turn in detail to the solutions of Eqs. (3.10) for different types of materials. Previously, the polarization dynamics of a single beam has been studied only for parity invariant isotropic media,[1,10] and for parity-invariant media with a C_4 rotation symmetry about the propagation axis. There, the approach was to apply the material symmetries present by deriving constraints on the form that the response tensors $\chi^{(3)}$ and η take. Here, we find it easier to impose any symmetry constraints directly on the material tensors $\mathbf{u}$, $\mathbf{v}$, and $\overleftrightarrow{\mathbf{w}}$ directly, by considering the resulting effect of symmetry operations in real space on tensors in Stokes-vector space. This allows us to establish a complete classification of the forms of $\mathbf{u}$, $\mathbf{v}$, and $\overleftrightarrow{\mathbf{w}}$ that are allowed for all rotation symmetries C_n about the propagation axis, for both parity-invariant and non-parity-invariant media (Table I). Proceeding through the different cases, we rely heavily on the rigid-body analogies of Sec. IV to help us find solutions. We are able to identify analytic solutions in terms of well-known functions for all cases where there is some rotation axis C_n, $n>2$, whether the material is parity invariant or not. This considerably extends what is known about the polarization dynamics of a single pulse or beam propagating through nonlinear media. We need not discuss any of the analytic solutions in detail once they are identified, since they are in fact familiar from the study of rigid-body dynamics.

Before beginning we mention that, as in Sec. IV, it is convenient to lump **u** and **v** together to write

$$\mathbf{b}\equiv-(\mathbf{v}+\mathbf{u}s_0)\ . \tag{5.1}$$

We note that since s_0 is arbitrary and a scalar in Stokes-vector space, symmetry constraints derived for **b** apply equally well to **v** (describing linear anisotropies) and **u** (describing self-induced anisotropies) separately.

We first consider a parity-invariant medium. Recall that the 2 component of the Stokes vector corresponds to circular polarization, while the 1 and 3 components to linear polarization. Thus, under parity inversion the 2 components go to minus themselves, while the 1 and 3 components are invariant,

$$(b_1,b_2,b_3)\to(b_1,-b_2,b_3)\ ,$$
$$\begin{bmatrix} w_{11} & w_{12} & w_{13} \\ w_{12} & w_{22} & w_{23} \\ w_{13} & w_{23} & w_{33} \end{bmatrix} \to \begin{bmatrix} w_{11} & -w_{12} & w_{13} \\ -w_{12} & w_{22} & -w_{23} \\ w_{13} & -w_{23} & w_{33} \end{bmatrix} . \tag{5.2}$$

So, for a parity-invariant medium, the most general form of the material tensors are

$$\mathbf{b}=(b_1,0,b_3),\ \ \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} w_{11} & 0 & w_{13} \\ 0 & w_{22} & 0 \\ w_{13} & 0 & w_{33} \end{bmatrix} . \tag{5.3}$$

That is [see Eqs. (3.11)], neither optical activity (linear or self-induced) nor linear-circular polarization interaction are allowed. We will return to this quite general case later, but first consider propagation along a symmetry axis. In real space the medium will then have some C_n, $n\geq 2$ rotation symmetry in the plane transverse to the propagation direction. A rotation of ϕ degrees about the propagation direction in real space corresponds to a rotation of 2ϕ degrees about the axis $\hat{2}$ in Stokes space. Thus, a C_{2n} (C_{2n+1}) symmetry in the transverse plane in real space corresponds to a C_n (C_{2n+1}) rotation symmetry about the 2 axis in Stokes space.

A real space symmetry C_2, therefore, corresponds to the identity operation in Stokes space, and there are no further constraints beyond the form (5.3). The real-space symmetry C_4 will be considered in a moment; we first discuss real-space symmetry C_3 and C_n, $n\geq 5$, which yield Stokes space symmetries C_n, $n\geq 3$, about the 2 axis. For these symmetries, which include complete isotropy, it

TABLE I. All of the $\overleftrightarrow{\mathbf{w}}$ tensors can be diagonalized by a rotation of the Stokes coordinate system without affecting the form of **b**.

Propagation-axis symmetries in real space		**b**	$\overleftrightarrow{\mathbf{w}}$
C_2	Nonparity invariant	(b_1,b_2,b_3)	$\begin{bmatrix} w_{11} & w_{12} & w_{13} \\ w_{12} & w_{22} & w_{23} \\ w_{13} & w_{23} & w_{33} \end{bmatrix}$
	Parity invariant	$(b_1,0,b_3)$	$\begin{bmatrix} w_{11} & 0 & w_{13} \\ 0 & w_{22} & 0 \\ w_{13} & 0 & w_{33} \end{bmatrix}$
C_4	Nonparity invariant	$(0,b_2,0)$	$\begin{bmatrix} w_{11} & 0 & w_{13} \\ 0 & w_{22} & 0 \\ w_{13} & 0 & w_{33} \end{bmatrix}$
	Parity invariant	(0,0,0)	$\begin{bmatrix} w_{11} & 0 & w_{13} \\ 0 & w_{22} & 0 \\ w_{13} & 0 & w_{33} \end{bmatrix}$
$C_3,C_n,n\geq 5$	Nonparity invariant	$(0,b_2,0)$	$\begin{bmatrix} w_{11} & 0 & 0 \\ 0 & w_{22} & 0 \\ 0 & 0 & w_{11} \end{bmatrix}$
	Parity invariant	(0,0,0)	$\begin{bmatrix} w_{11} & 0 & 0 \\ 0 & w_{22} & 0 \\ 0 & 0 & w_{11} \end{bmatrix}$

is easy to verify that the tensors (5.3) must reduce to the form

$$\mathbf{b}=(0,0,0),\quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} I_1^{-1} & 0 & 0 \\ 0 & I_2^{-1} & 0 \\ 0 & 0 & I_1^{-1} \end{bmatrix}, \tag{5.4}$$

where we are using the terminology of Sec. IV for the diagonal elements of $\overleftrightarrow{\mathbf{w}}$. The result can immediately be seen for $\mathbf{b}$, since for any $n>1$ a rotation of $2\pi/n$ about the 2 axis of a nonvanishing vector of the form in (5.3) would change the vector. For $\overleftrightarrow{\mathbf{w}}$, the matrices describing such a rotation must be applied to the second-rank tensor in (5.3); when the expressions are written out it is clear that for $n \neq 1$ or 2 the form in (5.3) can be invariant only if $w_{13}=0$ and $w_{11}=w_{33}$. On physical grounds one might not immediately expect that real-space symmetries C_3 and C_n, $n \geq 5$, should all exhibit the same polarization dynamics: Surely a C_3 real-space symmetry axis (which leads to C_3 in Stokes space) is distinguishable from, say, a C_8 real-space symmetry axis (which leads to a C_4 in Stokes space). But such distinctions would only appear through a fifth-order susceptibility, and yet higher distinctions would appear through higher-order nonlinearities. At our level of considering only a $\chi^{(3)}$ nonlinearity, all Stokes space symmetries about the 2 axis of the form C_n, $n \geq 3$ (including isotropy) lead to the same form (5.4). No anisotropies (linear or self-induced) are allowed, but circular-circular polarization interaction, and linear-linear polarization interaction described by one independent coefficient, are allowed [cf. Eqs. (3.11)]. Substituting Eqs. (5.4) into the equation of motion (3.10) gives Euler's equations of motion for a torque-free symmetric rigid body (two moments of inertia equal) which, as is well known,[23] can be trivially integrated in terms of trigonometric functions.

We now consider the special case of propagation along a C_4 symmetry axis in a parity-invariant medium. In Stokes-vector space this appears as a C_2 rotation symmetry about the $\hat{2}$ axis; the vector $\mathbf{b}$ must still vanish, but the second-rank tensor $\overleftrightarrow{\mathbf{w}}$ can take its full form (5.3) (see discussion after that equation). Thus we have

$$\mathbf{b}=(0,0,0),\quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} w_{11} & 0 & w_{13} \\ 0 & w_{22} & 0 \\ w_{13} & 0 & w_{33} \end{bmatrix}. \tag{5.5}$$

The tensor $\overleftrightarrow{\mathbf{w}}$ can be completely diagonalized by a simple rotation about the 2 axis; such a rotation corresponds, in real space, to rotating the x and y axes (transverse coordinates) about the z axis (propagation direction). That is, by merely redefining our linear polarizations we can diagonalize $\overleftrightarrow{\mathbf{w}}$ and obtain material tensors in the form

$$\mathbf{b}=(0,0,0),\quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} I_1^{-1} & 0 & 0 \\ 0 & I_2^{-1} & 0 \\ 0 & 0 & I_3^{-1} \end{bmatrix}. \tag{5.6}$$

No linear or nonlinear anisotropies are allowed, nor linear-circular polarization interaction, but circular-circular polarization interaction can be present, as well as two independent coefficients describing linear-linear polarization interaction. Thus, in contrast to C_3 and C_n, $n \geq 5$ symmetries in real space for which $I_1=I_3$, the "moments of inertia" I_1, I_2, and I_3 are, in general, unequal. Substitution of Eqs. (5.6) into the equations of motion (3.10) give Euler's equations of motion for a torque-free unsymmetric rigid body, which can be integrated in terms of elliptic functions.[23] The qualitatively new result that arises, over the problem of the symmetric body, is that rotation is unstable about the "middle axis," corresponding to the principal axis with the intermediate moment of inertia. It would be interesting to observe the optical analog of this effect.

Returning to the general Eqs. (5.3), which are the expressions for the material tensors that characterize an arbitrary propagation direction in a parity-invariant medium, we see that we can rotate our Stokes coordinate system about the 2 axis—corresponding to redefining our directions of linear polarization—to diagonalize $\overleftrightarrow{\mathbf{w}}$ and obtain material tensors in the form

$$\mathbf{b}=(b_1,0,b_3),\quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} I_1^{-1} & 0 & 0 \\ 0 & I_2^{-1} & 0 \\ 0 & 0 & I_3^{-1} \end{bmatrix}, \tag{5.7}$$

where, in general, I_1, I_2, and I_3 are unequal. The result is the same as for propagation along a C_4 real-space symmetry axis, except that linear and self-induced birefringence, each characterized by two independent components [cf. Eq. (5.1)] are allowed. Substitution of Eq. (5.7) into the equations of motion (3.10) gives

$$\begin{aligned} \dot{s}_1 &= -b_3 s_2 + (I_3^{-1} - I_2^{-1}) s_2 s_3 , \\ \dot{s}_2 &= b_3 s_1 - b_1 s_3 + (I_1^{-1} - I_3^{-1}) s_1 s_3 , \\ \dot{s}_3 &= b_1 s_2 + (I_2^{-1} - I_1^{-1}) s_1 s_2 . \end{aligned} \tag{5.8}$$

To solve (5.8) one uses the two invariants (3.12) to write two of the components of the Stokes vector as a function of the third. Then, in principle, one of the equations (5.8) can be integrated for the independent component.

We now discuss the yet more general problem of the polarization dynamics of light propagating through a non-parity-invariant medium. This problem is considered here for the first time. The argument of Eqs. (5.2) and (5.3) cannot be made, and for propagation along an axis not characterized by any particular symmetry we have material tensors of the greatest generality,

$$\mathbf{b}=(b_1,b_2,b_3),\quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} w_{11} & w_{12} & w_{13} \\ w_{12} & w_{22} & w_{23} \\ w_{13} & w_{23} & w_{33} \end{bmatrix}. \tag{5.9}$$

Before considering these forms, we look at the simplifications that result if the direction of propagation is characterized by a symmetry axis. For a real-space symmetry axis C_2—corresponding to the identity operation in Stokes-vector space—there is no further simplification. For a real-space symmetry axis of C_3 or C_n, $n \geq 5$, we have a Stokes space symmetry of C_n, $n \geq 3$ about the 2 axis, and the approach outlined after Eq. (5.4) leads to the forms

$$\mathbf{b}=(0,b_2,0), \quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} I_1^{-1} & 0 & 0 \\ 0 & I_2^{-1} & 0 \\ 0 & 0 & I_1^{-1} \end{bmatrix} . \tag{5.10}$$

This is the same as for a parity-invariant medium with the same propagation axis symmetry, except that linear and self-induced optical activity are allowed. Substituting Eq. (5.10) into the equations of motion (3.10) gives a Bloch equation

$$\dot{\mathbf{s}}=\mathbf{c}\times\mathbf{s} , \tag{5.11}$$

where

$$\mathbf{c}=(0,b_2+(I_1^{-1}-I_2^{-1})s_2,0) \tag{5.12}$$

is a constant vector. The Stokes vector then precesses about the $\hat{2}$ axis with an angular frequency of

$$|\,\mathbf{c}\,| = |\,b_2+(I_1^{-1}-I_2^{-1})s_2\,| \ . \tag{5.13}$$

In real space this precession corresponds to the rotation of the semimajor axis of the polarization ellipse, similar to but more general than the effect first predicted and observed for isotropic parity-invariant media by Maker, Terhune, and Savage[1] (In the case of 1, $b_2=0$). Two steady-state solutions of Eq. (5.11) are left- and right-handed circular polarizations.

For the special case of propagation along a C_4 real-space symmetry axis in a nonparity-invariant medium, we find that the material tensors (5.9) are constrained to take the form

$$\mathbf{b}=(0,b_2,0), \quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} w_{11} & 0 & w_{13} \\ 0 & w_{22} & 0 \\ w_{13} & 0 & w_{33} \end{bmatrix} , \tag{5.14}$$

again differing from a parity-invariant medium with the same propagation-axis symmetry by the presence of linear and self-induced optical activity [cf. Eq. (5.5)]. As in that case, by a rotation of the Stokes coordinate system about the 2 axis the $\overleftrightarrow{\mathbf{w}}$ in Eq. (5.14) can be diagonalized and we have material tensors in the form

$$\mathbf{b}=(0,b_2,0), \quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} I_1^{-1} & 0 & 0 \\ 0 & I_2^{-1} & 0 \\ 0 & 0 & I_3^{-1} \end{bmatrix} , \tag{5.15}$$

where in general I_1, I_2, and I_3 are not equal. Substituting Eqs. (5.15) into Eq. (3.10) gives the following equations of motion:

$$\begin{aligned} \dot{s}_1 &= b_2 s_3 + (I_3^{-1}-I_2^{-1})s_2 s_3 , \\ \dot{s}_2 &= (I_1^{-1}-I_3^{-1})s_1 s_3 , \\ \dot{s}_3 &= -b_2 s_1 + (I_2^{-1}-I_1^{-1})s_1 s_2 . \end{aligned} \tag{5.16}$$

These equations are formally equivalent (under a permutation of 1, 2, and 3) to those equations describing a dc-field-induced birefringence, which have been studied and solved.[10,11] Hence that analysis also applies to Eqs. (5.16).

Finally, we consider propagation along an arbitrary direction in a non-parity-invariant medium. In this case the material tensors take their full form (5.9), although we can still diagonalize $\overleftrightarrow{\mathbf{w}}$ by a rotation in Stokes-vector space, yielding expressions of the form

$$\mathbf{b}=(b_1,b_2,b_3), \quad \overleftrightarrow{\mathbf{w}}=\begin{bmatrix} I_1^{-1} & 0 & 0 \\ 0 & I_2^{-1} & 0 \\ 0 & 0 & I_3^{-1} \end{bmatrix} . \tag{5.17}$$

Note that here, of course, the rotation will not necessarily be about the 2 axis. Used in the equations (3.10), Eqs. (5.17) lead to the most complicated form the Stokes parameter equations take,

$$\begin{aligned} \dot{s}_1 &= b_2 s_3 - b_3 s_2 + (I_3^{-1}-I_2^{-1})s_2 s_3 , \\ \dot{s}_2 &= b_3 s_1 - b_1 s_3 + (I_1^{-1}-I_3^{-1})s_1 s_3 , \\ \dot{s}_3 &= b_1 s_2 - b_2 s_1 + (I_2^{-1}-I_1^{-1})s_1 s_2 . \end{aligned} \tag{5.18}$$

Nonetheless, there are still two invariants of the motion (3.12), and using them to eliminate two of the variables leaves in principle the task of integrating only one equation of motion for the remaining component.

This completes the classification of the polarization dynamics of a beam, or pulse, propagating through a nonlinear medium. The results for different propagation-axis symmetries, for parity-invariant and non-parity-invariant media, are summarized in Table I. We emphasize that for cases where any propagation-axis symmetry C_n, $n>2$ is present, we have identified an analytic solution in terms of well-known functions, for both parity-invariant and non-parity-invariant media. Even if such a symmetry is not present, the two general invariants of Eq. (3.10) can be used to reduce the problem of polarization dynamics [Eqs. (5.8) or (5.18)] to one of quadrature.

VI. DISCUSSION

The usefulness of the Stokes-vector formalism for studying nonlinear polarization dynamics has been noted before,[6–11] but for the first time we have here exploited the full SO(3) covariance of the Stokes parameters to present a classification of the solutions that result for pulse propagation in parity-invariant and non-parity-invariant media, with arbitrary propagation-axis rotation symmetries. Thus, unlike earlier workers who have dealt only with parity-invariant isotropic media, and propagation along an axis with rotational symmetry C_4 in a parity-invariant media, our equations, in general, describe a host of physical effects (such as linear and self-induced optical activity) not previously considered in the single pulse or beam propagation problem.

In the most general case, the problem is formally equivalent to the airplane-and-rotor problem of rigid-body dynamics. We have proved the general existence of a second invariant, which reduces the solution of the polarization dynamics to quadrature, and have discovered the physical significance of that invariant. Further, in all but the most complicated cases we have identified analytic solutions of the polarization dynamics in terms of well-known functions.

Many extensions of this work are possible. The inclusion of group-velocity dispersion would make the pulse calculation presented here more applicable to the ultrashort pulses that can now be generated. With the benefit of hindsight, one can see that the Stokes-vector equation of motion (3.10) could have been written down immediately, upon the assumptions of SO(3) covariance and constant energy flux s_0. In such a manner, higher-order nonlinearities can easily be included. The generalization to the interaction of two counterpropagating beams is a problem we turn to in a future communication; we also plan to consider in detail the effect of dc-field-induced birefringence on the polarization dynamics of a pulse. This latter phenomenon is, of course, of interest with respect to possible applications in optical devices.[24,25] Although these topics have been discussed before in special cases, using either the electric field amplitude formalism[24] or Stokes parameters[10,11] the full SO(3) covariance of the Stokes vector has not been applied. On the basis of this work, its use can be expected to considerably simplify and clarify those problems, highlighting the points of physical significance, as well as allowing the treatment of more general symmetries.

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